A BIOTECHNOLOGICAL APPROACH TO MITIGATE GREEN HOUSE GAS EMISSIONS FROM COAL MINE VENTILATION AIR IN AUSTRALIA

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Abstract

Coal mines are the second largest contributor of anthropogenic methane (CH_4) emissions with an estimated emission of around 5-30 Tg CH_4 year^{-1}. Considering the global warming potential of CH_4 (~ 25 times that of CO_2 over a 100 year period), developing a unique technology to mitigate the low levels of CH_4 (0.2-5 %) emissions especially coming from coal mine ventilation air is highly important for countries with considerable coal reserves like Australia. A research project "***Bioremediation of methane from mine ventilation air***" jointly funded by the Advanced Manufacturing Cooperative Research Centres (AMCRC) and MBD Energy Ltd., Australia, aims to develop a dual culture system to convert CH_4 into green fuels using indigenous bacteria and cyanobacteria in novel bioreactors. Methanotrophic bacteria are capable of metabolizing CH_4 into CO_2 under optimized bioreactor conditions. Subsequently, the CO_2 can be converted into oxygen (O_2) by cyanobacteria resulting in biomass generation for biofuel production. This study is the first of its kind in developing such novel bioreactors to implement at commercial sites after vigorous laboratory testings/optimization to mitigate both CH_4 and CO_2. Further, this technology is readily transferable to other CH_4 generating industries like landfills, anaerobic composting systems and dairy farms. In addition, successful implementation allows claiming of Clean Development Mechanism (CDM) benefits for carbon capturing in Australia and other parts of the world.

Introduction

Use of fossil fuels has led to unprecedented global surface warming. According to the NOAA Satellite and Information Services, global surface temperature increased by 0.54 °C above the long-term century average during the period 2000-2009, whereas in 1990, 'only' a 0.36 °C rise was predicted [1]. It is projected that warming will still increase in the next decade. Of the three fossil fuels (coal, petroleum, natural gas), coal reserves are most widely distributed in over 100 countries on all continents (except Antarctica) and Australia was the major coal exporter in 2009 with 288.5 Mt [2]. Methane is released into the atmosphere by natural and anthropogenic activities. Methane release by industries is highest for the oil and gas industry, with landfill and coal mining emissions being in 3rd and 6th position on the atmospheric contribution scale, respectively [3].

Methane (CH_4) is a greenhouse gas (GHG) with a 100 year warming potential 21 times that of CO_2 [3] although it was upgraded to 25 in 2007 in the 4^{th} report on climate change by the Intergovernmental Panel on Climate Change [4]. CH_4 is trapped between the hard rocks during the geological formation of coal, it is released during mining operations. While coal-mining-derived CH_4 is also being used to generate electricity, 90% of coal mines release CH_4 directly into the atmosphere, as current extraction processes do not allow for a 100% recovery with 70% of all coal-mining-associated methane being emitted as mine ventilation air methane (VAM) [5].

VAM 'only' contains 0.2-1% CH_4, with an average VAM content of 0.65% reported. CH_4-levels above 5% threaten mine safety due to explosion risk resulting in costly mine evacuations. For example, large air flows (with ~440 m^3 s^{-1} reported for one Australian mine) are required to maintain levels below the explosive threshold, resulting in an average annual release of 18 - 60 kt of CH_4 with a CO_{2eq} of 0.5 – 1.5 Mt per mine ventilation shaft for these concentrations and air flows (IPCC 25x multiplier). Published ventilation air flows for averaged-sized coal mines, producing 1 Mt of coal per year, are lower at 167 m^3 s^{-1} with an average of 0.3% CH_4, which nonetheless still results in an average CO_{2eq} output of 0.26 Mt per annum [6].

Currently, no technology exists to capture CH_4 present at concentrations below 1% economically, but predicted global warming and the increasing carbon-constraint of global economies demands solutions to GHG emission. This is particularly important for the large Australian coal mining sector, which in the FY 2009-2010 mined 359 Mt of black coal of which 113 Mt were produced through underground mining generating a total income of > $35 billion nationwide and employing more than 26,000 people in Queensland alone [7]. Given the long-term socioeconomic and economic importance of Australian coal mining, introducing clean coal technologies will lower GHG emissions and guarantee long-term viability of this sector of the mining industry, particularly in rural Queensland.

GHG remediation strategy

Any mitigation strategy has to be ultimately economically and environmentally sustainable. Therefore our research project aims to implement and test novel methanotrophic biofilters and diazotrophic cyanobacteria cultivation for CH_4 and CO_2 remediation from mine ventilation air whilst generating value-adding co-products, such as biofuels and biopolymers from the biomass generated in both stages of the remediation approach (Fig.1). In this novel approach, a packed-bed trickling biofilter colonised with methanotrophic consortia converts CH_4 to CO_2 and a hanging biofilm diazotrophic (capable of atmospheric nitrogen assimilation) cyanobacteria cultivation system converts mine CO_2 and methane-derived CO_2 to oxygen and biomass.

Conversion of CO_2 by diazotrophic cyanobacteria was chosen to save on nitrogen fertilisation costs, but any microalga or non-nitrogen fixing cyanobacterium is suitable, provided it is capable of biofilm formation. We designed the novel biofilm-based cultivation system, as it saves on costs and energy required with the dewatering (harvest) of the biomass [8]. Dewatering is the largest single factor impacting on the economical and environmentally sustainable production (mainly energy and therefore carbon and area footprint of the industry) of low-value bio-products such as aviation and biofuels markets. The surplus biomass of both systems is then converted to value-adding bio-products, with product value being inversely correlated with market size (i.e. high value bio-products (+) have a restricted market size (-)) (Fig. 1).

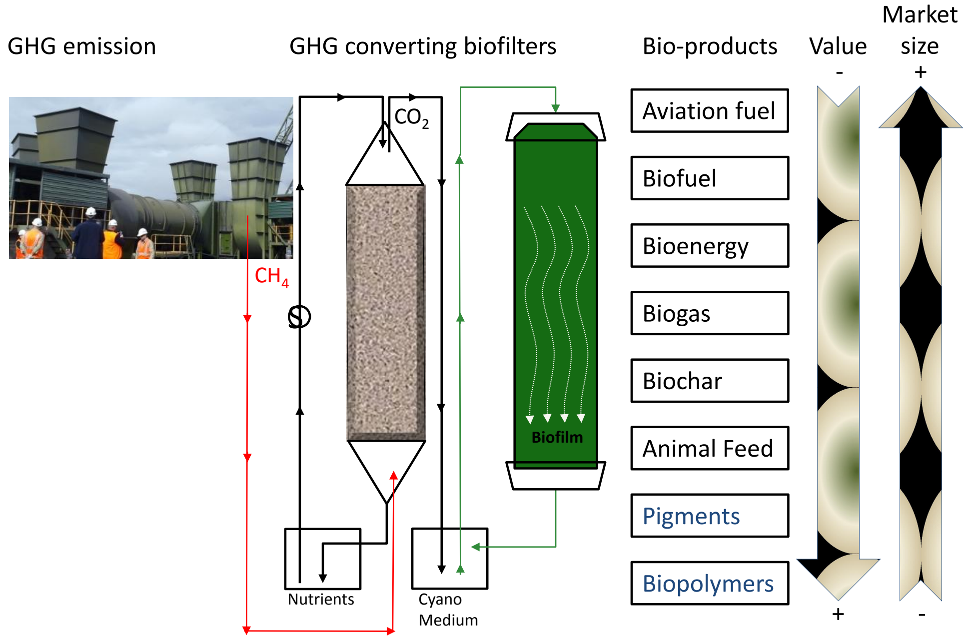

Fig.1: Process diagram for carbon mitigation using novel methanotrophic biofilters in conjunction with diazotrophic cyanobacterial cultivation for CH_4 and CO_2, respectively, whilst generating value-adding co-products.

The choice of methanotrophic bacteria is less clear cut, as the selection needs to be based on 1. Methane oxidation efficiencies and 2. Biomass potential for bio-product development. In general, type II methanotrophs have higher biomass application potentials (other than for direct energy generation and biochar), because, both, the presence of oleic acid (C18:1) together with the ability to store methane-derived carbon in the form of poly(3-hydroxyl)butyrate (PHB) (Table 1), makes them ideal candidates for biopolymer production (see below). In addition, the ability to assimilate atmospheric nitrogen would save on nitrogen fertilisation costs, however, type I methanotrophs may have a positive effect on the methane oxidation capacity of a biofilter (increasing carbon tax savings) and also could influence type II methanotrophs through positive allelopathic interactions when cultivated in a mixed species consortium within the biofilter.

Economic benefits

Modelling of economics have to take direct first and second order income and expenditure scenarios into account, which therefore requires a multifactorial approach. A third order, which should be considered, is the potential income generated by technology transfer, as the proposed remediation systems are immediately applicable to other methane-emitting industries, particularly landfills, globally the 3rd largest emitter of CH_4. As the above GHG remediation strategy is in the conceptual/laboratory testing phase, only carbon-tax-based expenditures, as well as rough estimates of biofilter capital and maintenance costs are possible. These costs are being offset by biomass-derived income, modelling only those that are of higher value to give a weighting on strategies and priorities of implementation and pathway to market developments.

Table 1: Taxonomic affiliation and critical characteristics of methanotrophic bacteria for cultivation and biopolymer production

Selection Character	Type I	Type X	Type 2
Taxonomic affiliation	Gamma-proteobacteria	Gamma-proteobacteria	***Alpha-proteobacteria***
Phylogenetic signature sequences	3	No	**1**
Major PUFAs	14.0, 16:1ω7c, 16:1ω5t	16:0, 16:1ω7c	***18:1ω8c***
Temperature tolerance	<30°C	<30°C	<30°C
Nitrogen provision			***Aerobic N_2 fixation***
PHB storage	No		***Yes***

First order income and costs relate to predicted carbon tax savings (income) and liabilities, as well as infrastructure/maintenance requirements (costs), whereas second order economics include product income (for both the methanotrophic biofilter biomass and the diazotrophic cyanobacterial biomass), which will offset carbon tax liabilities, infrastructure/maintenance costs and is expected to generate net long-term income.

Generated biomass offers the opportunity to develop the large non-petrochemical-based fuel and plastic markets, additionally offsetting Australia's GHG emissions relating to their import (transportation) and present day petrochemical-based production further. Hence this proposed project will make a significant positive contribution to the public perception of the Australian coal mining industry as an innovative, environmentally and economically sustainable industry sector. In addition, large-scale bio-product industries would need to be built in close proximity to underground coal mines or other methane-emitting industries. These industries themselves offer new employment opportunities in regional/rural areas and hence have their own socio-economic benefits.

Carbon tax liabilities

Carbon tax liabilities were modelled for averaged-sized coal mines, producing 1 Mt of coal per year, with air flows of 167 $m^3 s^{-1}$ and an average methane concentration of 0.3% v/v. Mine ventilation air concentrations were calculated based on weight percent of CH_4 in air at 20 °C, at sea level and standard atmospheric pressure resulting in an average CO_{2eq} output of 0.26 Mt per annum [6]. The recommended CO_{2e} factor of 19.1 and a tax of \$23 t^{-1} CO_{2e} was applied resulting in an annual carbon tax liability of ~6 million AUD.

Carbon tax savings, will depend on how much volume of the total air flow per ventilation shaft can be oxidised to CO_2 using the proposed technology.

Methanotrophic biofilter costs
Biofilter performance is influenced by temperature, methane concentration and retention time of the feed gas stream, and will other than that depend in principal on two factors 1 & 2, which link to additional factors and dictate performance:

1: the materials selected for the biofilters *per se* and
2. the oxidation efficiency of the methanotrophic bacteria colonising the biofilter material (e.g. different strains or consortia of different species will have their individual maximal CH_4 oxidation rate based on enzyme kinetics in the oxidation pathway),
3. the concentration of methanotrophs per unit volume, which is directly linked to bed material choice (e.g. pore size), which in turn will influence
4. the air flow rate maxima, where the biofilters can perform optimally.

These factors will govern the capital expenditure for construction of the biofilters (e.g. number and cost of systems that need to be applied for 100% CH_4 and resulting CO_2 remediation).

Annualised capital costs of a methanotrophic biofilter was taken from Yoon et al. [9], who modelled biofilter performance using available enzyme kinetics for *Methylosinus trichosporium* and air flow and packaging materials from biofilters installed and operational at landfills. The costs, which, according to the authors, include capital, operational and maintenance costs were $80,000 per biofilter per year for systems with a diameter of 3.66 m and a height of 11.5 m, which are the likely dimensions for the methanotrophic biofilters for the proposed project.

Bio-product-derived income streams
Total income potential assumes pigment extractions are carried out first and the left over biomass is used for non-pigment products such as animal feeds, biochar (excluding resulting pyrolysis biocrude as a co-product), or aviation fuel (biofuels). As the sale of the later three products is mutually exclusive, Totals I, II and III, incorporate only one of the respective products (Table 2).

Methanotrophs
Biofilter bed materials are colonised by methanotrophs and organism growth will ultimately lead to filter clogging. Hence biofilters need to be flushed on a regular basis to remove this biomass. The normally wasted biomass, can be to put to good use, by switching cultivation to nitrogen limiting conditions prior to flushing to allow for the accumulation of poly-3-hydroxybutyrate (PHB), a polyhydroxyalkanoate (PHA) with properties similar to polypropylene. PHB in general is a microbial product and accumulation between 40-80% of biomass dry weight has been reported [10]. PHB is non-toxic and is a key ingredient in the biodegradable plastic industry and also in the biomedical market (internal sutures).

Reported market price for PHB ranges from $4.4 - 16.25 kg^{-1} [11,12]. Calculating annual income potential from this ‘waste’ methanotroph biomass would require detailed knowledge on biofilm colonisation density (e.g. g dry weight per unit area or volume) and CH_4 abatement and kinetics thereof, which is strongly temperature and CH_4-input sensitive. We have been unable to acquire such parameters from published data [13], but it can be reasonably assumed that tons of methantroph biomass needs to be maintained for efficient and effective remediation of CH_4. To approximate an income, the following case scenario was set: An annual biomass of 45 t for extraction of PHB with a content of 40% and a market price for the extracted PHB of 8 AUD kg^{-1}. Taking into consideration that a bio-plastic industry could be co-localised with underground coal mines, a second scenario with the same biomass assumptions but the actual market prize of 160 AUD kg^{-1} for the finished PHB pellets has also been presented (Table 2). No other methanotrophic biomass-derived bio-product, such as biochar etc. has been included. Based on the case parameters set, PHB production by methanotrophic bacteria can offset carbon tax liabilities by ~144K to $2.88M AUD, respectively annually.

Table 2: Estimated project incomes in AUD based on 10% methane remediation capacity and the generation of value-adding co-products using methanotrophic in conjunction with diazotrophic cyanobacterial biofilters

Costs			
Carbon tax liabilities		$6,000,000	
Income			
Carbon tax savings			
	CH_4-derived[aa]	$600,000	
Biomass-derived			
Methanotrophs		***PHB unprocessed (AUD yr^{-1})***	***PHB pellets (AUD yr^{-1})***
	PHB[a]	$144,000	$2,880,000
Cyanobacteria		***suspension culture[1] (AUD ha^{-1} yr^{-1})***	***biofilm cultivation[2] (AUD ha^{-1} yr^{-1})***
	Phycocyanin[b]	$1,050,000	$1,050,000
	other pigments (Astaxanthin)[c]	$603,750	$60,375,000
	animal feed[d]	$35,000	$3,500,000
	biochar[e]	$21,000	$2,100,000
	aviation fuel[f]	$32,012	$3,201,220
Income generated			
Total I (animal feed)		***2,432,750***	***67,355,000***
Total II (biochar instead of animal feed)		***2,418,750***	***67,005,000***
Total III (aviation fuel instead of animal feed)		***2,429,762***	***68,106,220***

[aa]assuming an initial 10% capacity for complete methane oxidation. [a]assuming a doubling time of six weeks, a harvest of 5 t every six weeks and a PHB content of 40%. [b]assuming a phycocyanin content of 5% at a price of $3 mg^{-1} kg^{-1} pure compound in a traditional suspension cultivation system (=500 kg pure compound). [c]Astaxanthin production was modelled on reported 0.35% dry biomass content produced on a one ha site equipped with RPM-PBRs. [d]animal feed production was also modelled on RPM-PBR-produced biomass on a one ha site, after pigment extractions (phycocyanin and Astaxanthin) with an estimated price of $500 t^{-1}. [e] & [f] Biomass assumptions are as for [d] assuming a 30% conversion efficiency and price estimates are $300 t^{-1} (not including income based on pyrolysis crude oil, a byproduct of the conversion process) and $1.25 L^{-1}. Total I, II, and III reflect the income generated by including either animal feed, biochar or aviation fuel, respectively.

Cyanobacteria

Biomass production was calculated on industrially achieved long-term averages at 20 g dry weight (DW) m^{-2} day^{-1}, although maximum productivities of 31 and 35 g m-2 day-1 in outdoor cyanobacterial bioreactors [14,15]. For the new biofilm cultivation system, for which a provisional patent application has been filed [8], a 100x biomass concentration factor has been applied, even though pilot studies suggest that the systems can achieve biomass concentration factors of >200x, compared to suspension systems using the same organisms and operated under the same climatic conditions [8]. Other than two order of magnitude improved production, the system offers significant energy and OPEX/CAPEX savings, as harvesting only requires scraping off the biomass from the solid support and no dewatering using costly and energy intensive dewatering technologies (e.g. Evodos centrifugation or filtration) is required. These savings have not been modelled, as there is insufficient data at present.

Phycocyanin:
Phycocyanin has large market potential due to a number of bioactive effects like antioxidant, anti-inflammatory, anti-artheriosclerosis, anti-carcinogenic and hepatoprotective actions (reviewed in [16]. Markets for these applications however, need to be developed. Prices listed for fluorescent tagging of compounds in biomedical research range from $3 – 17 per mg of the pure pigment and 5 – 10% content per biomass dry weight can be achieved by selecting appropriate cultivation conditions. Phycocyanin contents of 11-17% have been reported for many diazotrophic cyanobacteria [17]. The current market volume for using phycocyanin for tagging in biomedical research is estimated to be $5 - 10M.

A content of 5% per biomass dry weight with the value restricted to $3 per mg pure compound was applied using a traditional suspension cultivation system. Likewise, the production was restricted to 1 t dry biomass (500 kg pure compound) per year, due to the current market size for tagging purposes; however, production can be easily up-scaled when other medical markets become available.

Given the above, the annual income generated would be *10.5 M AUD for the production of 500 kg pure compound requiring 1 t biomass dry weight production* (Table 2), which can be achieved on one seventh of a ha in standard suspension systems or 15 m^2 production area, even when using a traditional suspension cultivation system instead of the more productive biofilm system designed by Berner and Heimann are used [8].

Other carotenoids:
Assuming pigment concentrations (0.35% of dry biomass) and restricting production to 1 ha, income generated from the remaining biomass would be *0.6 or 60 M AUD* for the cultivation systems under consideration at $2,500 per kg of the pure compound (Table 2).

Other products:
After pigment extraction, the biomass can be used to either produce animal feed, if there is a market close by, or biochar or aviation fuel. Again production has been restricted to 1 ha and the two different cultivation systems have been used in these calculations (Table 2).

Animal feed can generate an additional income of *$35K or $3.5M* per respective cultivation system at an assumed market price of $500 per t DW, however, a close by market option is essential.

Biochar has applications as a soil conditioner and at $300 per t DW could generate $21K and $2.1M, respectively, should animal feed production turn out to be not feasible, either due to the biochemical profile of the biomass produced or no direct access to the market.

In contrast, biofuels are a much cheaper commodity, but markets are unsaturable. Based on a 30% conversion efficiency using hydrothermal liquefaction HTL (Solray pers. com.) and applying the specific gravity of ~0.82 L^{-1}, calculated incomes would be $32K and $3.2M, respectively at that scale, producing 26 to 2600 kL of aviation fuel.

Conclusions

Co-installation of methanotrophic biofilters with a cyanobacterial production system taking the CO_2 off gas of the biofilters can turn a tax liability into a profitable business through generation of high-value commodities even on a restricted 1 ha site. Particularly the co-localisation of bioplastic producing industries with underground coal mining, offers clear benefits for PHB production from methane and efficient and cost-effective processing, negating carbon-negative transport of raw materials from rural Australia to coast-based industries. Cultivation system innovation for the cyanobacterial biomass is a clear driver for profit compared to traditional suspension culture systems.

It is apparent that the main income drivers, capable to offset the installation and operation costs are not projected carbon tax savings, as it is likely that a staged installation plan would be required. The high value PHB obtained through methane oxidation by methanotrophic consortia, the phycocyanin market and installation of the biofilm-based cyanobacterial cultivation system instead of the traditional suspension culture systems improves income balances considerably. In particular the biofilm-based cultivation of the diazotrophic cyanobacterial biomass improves areal productivities by two orders of magnitude, even though savings of capital and operational expenditures of the system itself and significantly simplified dewatering technologies [8] were not considered, as there are insufficient data for industrial-scale operation.

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